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### (54) METHOD AND APPARATUS FOR CONVERTING HYDROCARBON FUEL INTO HYDROGEN GAS AND CARBON DIOXIDE

VERFAHREN UND VORRICHTUNG ZUR UMSETZUNG VON  
KOHLENWASSERSTOFFBRENNSTOFF IN WASSERSTOFF UND KOHLENDIOXYD

PROCEDE ET APPAREIL POUR TRANSFORMER DU CARBURANT HYDROCARBURE EN GAZ  
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**Description****Background of the Invention**

**[0001]** Fuel cells continue to play an increasingly important role in power generation for both stationary and transportation applications. A primary advantage of fuel cells is their highly efficient operation which, unlike today's heat engines, are not limited by Carnot cycle efficiency. Furthermore, fuel cells far surpass any known energy conversion device in their purity of operation. Fuel cells are chemical power sources in which electrical power is generated in a chemical reaction between a reducer (hydrogen) and an oxidizer (oxygen) which are fed to the cells at a rate proportional to the power load. Therefore, fuel cells need both oxygen and a source of hydrogen to function.

**[0002]** There are two issues which are contributing to the limited use of hydrogen gas today. Firstly, hydrogen gas ( $H_2$ ) has a low volumetric energy density compared to conventional hydrocarbons, meaning that an equivalent amount of energy stored as hydrogen will take up more volume than the same amount of energy stored as a conventional hydrocarbon. Secondly, there is presently no widespread hydrogen infrastructure which could support a large number of fuel cell power systems.

**[0003]** An attractive source of hydrogen to power fuel cells is contained in the molecular structure of various hydrocarbon and alcohol fuels. A reformer is a device that breaks down the molecules of a primary fuel to produce a hydrogen-rich gas stream capable of powering a fuel cell. Although the process for reforming hydrocarbon and alcohol fuels is established on a large industrial basis, no known analogous method for converting an alcohol or hydrocarbon fuel into hydrogen gas and carbon dioxide.

**[0004]** EP-A-0600621 discloses a combined reformer and shift reactor comprising a cylindrical reforming chamber arranged within, and on the axis, of a cylindrical vessel. An annular steam generator is arranged within, and coaxially with, the vessel. The steam generator is arranged around the reforming chamber. A plurality of shift reactors extend axially, with respect to the vessel through the steam generator. Methane and steam are supplied via a helically coiled pipe to the reforming chamber and air is supplied via a helically coiled pipe. The methane and steam mixture and air flowing through the pipes are preheated by the reforming chamber product gases. The shift reactors convert carbon monoxide and water in the product gases to carbon dioxide and hydrogen and the heat produced is transferred to the water in the steam generator to produce steam.

**[0005]** According to one aspect of the present invention there is provided a reformer for converting a hydrocarbon fuel into hydrogen gas and carbon dioxide comprising:

a first vessel having a first portion defining a partial

oxidation reaction zone and a first vessel inlet adapted to direct reactants including oxygen-containing gas and unburned fuel tangentially into the first portion of the first vessel and, a first vessel outlet disposed downstream of the partial oxidation reaction zone.

**[0006]** In an embodiment of the invention, the reformer includes a first vessel having a partial oxidation reaction zone and a separate steam reforming reaction zone. The first vessel has a first vessel inlet at the partial oxidation reaction zone and a first vessel outlet at the steam reforming zone. The reformer zone also includes a helical tube extending about the first vessel. The helical tube has a first end connected to an oxygen-containing source and a second end connected to the first vessel at the partial oxidation reaction zone. Oxygen gas from an oxygen-containing source can be directed through the helical tube to the first vessel. A second vessel having a second vessel inlet and second vessel outlet is annularly disposed about the first vessel. The helical tube is disposed between the first vessel and the second vessel and gases from the first vessel can be directed through the second vessel.

**[0007]** According to a second aspect of the invention there is provided a method for converting hydrocarbon fuel into hydrogen gas and carbon dioxide within a reformer comprising the steps of:

a. directing a stream including an oxygen-containing gas adjacent to a first vessel to pre-heat the oxygen-containing gas by heat transfer from the first vessel;

b. introducing a stream including unburned fuel into the oxygen-containing gas stream to form a mixture including oxygen-containing gas and fuel;

c. directing the mixture of oxygen-containing gas and fuel tangentially into a partial oxidation reaction zone within the first vessel; and

d. directing the mixture of oxygen-containing gas and fuel through the partial oxidation reaction zone to produce a heated reformate stream including hydrogen gas and carbon monoxide.

**[0008]** An embodiment of the method of the invention includes directing oxygen-containing gas through a helical tube which is disposed around a first vessel. Hydrocarbon vapor and steam are directed into the helical tube to form a mixture of oxygen gas, fuel vapor and steam. The mixture of oxygen gas, fuel vapor and steam are directed into the first vessel. The fuel vapor spontaneously partially oxidizes to form a heated reformate stream that includes carbon monoxide and hydrogen gas. The remaining fuel vapor is steam reformed in the heated reformate stream to form hydrogen gas and carbon monoxide. The heated reformate stream is directed over the exterior of the helical tube, whereby the heated reformate stream heats the mixture in the helical tube.

A portion of the carbon monoxide gas of the reformate stream is converted to carbon dioxide and hydrogen gas by a high temperature shift reaction. At least a portion of the remaining carbon monoxide gas of the reformate stream is converted to carbon dioxide and hydrogen gas by a low temperature shift reaction.

**[0009]** This invention has many advantages. The apparatus can use a variety of hydrocarbon fuels, such as gasoline, JP-8, methanol and ethanol. The partial oxidation reaction zone allows the fuel to partially burn while not forming soot and while providing heat to the steam reforming zone and the other portions of the reactor annularly disposed around the partial oxidation zone. Further, the apparatus is sufficiently compact for use in an automobile. In some embodiments, the apparatus includes a high temperature shift catalyst which allows the apparatus to be more compact and lighter in weight than if only a low temperature shift catalyst is used.

#### Brief Description of the Drawings

**[0010]** The invention will now be described by way of example only with reference to the accompanying drawing, the sole Figure of which is an orthogonal projection side view of one embodiment of the apparatus of the present invention.

#### Detailed Description of the Invention

**[0011]** It will be understood that the particular embodiment of the invention is shown by way of illustration without limiting the scope of the invention. The principal features of this invention can be employed in various embodiments without departing from the scope of the invention. All percentages and parts are by weight unless otherwise indicated.

**[0012]** Referring to the drawing, reformer 10 has reformer vessel 12. Reformer vessel 12 can be cylindrical in shape. Reformer 10 has upper portion 14 and lower portion 16. Disposed in the center of reformer vessel 12 is first vessel 18 which extends substantially the height of reformer vessel 12. First vessel 18 has first vessel inlet 20 for receiving gases into first vessel 18 and can tangentially direct the gases through the first vessel. First vessel 18 has first vessel outlet 22 at upper portion 14 of reformer 10 for gases to exit first vessel. Perforated plate 31 is located at first vessel outlet 22 and covers the diameter of first vessel 18. Partial oxidation reaction zone 24 is in lower portion 16 of first vessel 18.

**[0013]** Partial oxidation zone 24 is suitable for partial oxidation of a hydrocarbon or alcohol fuel with oxygen to form a mixture including carbon monoxide, steam and hydrogen gas. Steam reforming zone 26 is above partial oxidation zone 24 and includes a steam reforming catalyst 28. Preferably, the steam reforming catalyst includes nickel with amounts of a noble metal, such as cobalt, platinum, palladium, rhodium, ruthenium, irid-

ium, and a support such as magnesia, magnesium aluminate, alumina, silica, zirconia, singly or in combination. Alternatively, steam reforming catalyst 28 can be a single metal, such as nickel, supported on a refractory

5 carrier like magnesia, magnesium aluminate, alumina, silica, or zirconia, singly or in combination, promoted by an alkali metal like potassium. Steam reforming zone 26 can autothermally reform steam and methane generated in partial oxidation zone 24 to hydrogen gas and carbon monoxide. Steam reforming catalyst 28, which can be granular, is supported within partial oxidation zone 24 by perforated plate 30 and perforated plate 31.

**[0014]** Helical tube 32 extends about the length of first vessel 18. First end 34 of helical tube 32 is located at inlet housing 33. Oxygen source 42 is connected to inlet housing 33 by conduit 35 with first end inlet 36 for receiving oxygen-containing gas from oxygen gas zone 40. Second end 44 of helical tube 32 is connected at first vessel inlet 20. Examples of suitable oxygen-containing gas include oxygen ( $O_2$ ), air, etc. Fuel inlet 46 is joined to helical tube 32 proximate to second end 44. Conduit 50 extends from fuel source 48 to fuel inlet 46. Examples of suitable fuels include hydrocarbons which encompass alcohols, also. Fuels include gasoline, kerosene, JP-8, methane, methanol and ethanol. Steam inlet 52 is proximate to fuel inlet 46. Steam can be directed from steam source 54 to steam tube 56 through first steam inlet 52 into helical tube 32. In another embodiment, fuel and steam can be directed into helical tube 32.

**[0015]** Second vessel 58 is annularly disposed about first vessel 18. Second vessel inlet 60 receives gaseous products from first vessel outlet 22. Second vessel outlet 62 at lower portion 16 of reformer 10 allows gas to exit second vessel 58. Helical tube 32 is disposed between first vessel 18 and second vessel 58 and gases from first vessel 18 can be directed through second vessel 58 from second vessel inlet 60 over and around helical tube 32 to second vessel outlet 62. Flow distribution region 63 conducts gas from second vessel outlet 62 to high temperature shift zone 64. Additional steam or water can be directed from a steam source into second vessel 58 through second steam inlet 53 to provide added steam to provide added cooling and further the reformation of the fuels.

**[0016]** High temperature shift zone 64 is annularly located between second vessel 58 and reformer vessel 12 and includes a high temperature shift catalyst. An example of a suitable high temperature shift catalyst are those that are operable at a temperature in the range of between about 300°C and about 600°C. Preferably the high temperature shift catalyst includes transition metal oxides, such as ferric oxide ( $Fe_2O_3$ ) and chromic oxide ( $Cr_2O_3$ ). Other types of high temperature shift catalysts include iron oxide and chromium oxide promoted with copper, iron silicide, supported platinum, supported palladium, and other supported platinum group metals, singly and in combination. High temperature shift catalyst

66 is held in place by perforated plate 68 and perforated plate 70. Gas can pass through high temperature shift zone 64 through perforated plate 70 to sulfur removal zone 71.

[0017] Above high temperature shift zone 64 is sulfur removal zone 71. Sulfur removal zone 71 includes a catalyst which can reduce the amount of hydrogen sulfide ( $H_2S$ ), which is deleterious to a low temperature shift catalyst, in the gas stream to a concentration of about one part per million or less. An example of a suitable catalyst includes a zinc oxide. Sulfur removal zone 71 is sized depending on the type of fuel used. If a low sulfur fuel is used, a small sulfur removal zone is needed. If a high sulfur fuel is used, a larger sulfur removal zone is necessary. Gas can pass from sulfur removal zone 71 through perforated plate 73 to cooling zone 72.

[0018] Cooling zone 72 includes a plurality of vertical fins 74 which radiate from second vessel 58 to reformer vessel 12 which extends from high temperature shift zone 64 to low temperature shift zone 76.

[0019] Cooling tube 78 is helically disposed about second vessel 58 and is attached to vertical fins 74. Cooling tube 78 has cooling tube inlet 80 for receiving a cooling medium, such as water, through cooling tube 78 to cooling tube outlet 82. In another embodiment, cooling tube 78 is wound a second series of times around second vessel 58. The gaseous products from high temperature catalyst zone 64 can pass between the vertical fins 74 and pass over cooling tube 78 allowing gaseous products to cool.

[0020] Low temperature shift zone 76 is annularly disposed above cooling zone 78 and between second vessel 58 and reformer vessel 12 and includes low temperature shift modifying catalyst 84 for reducing carbon monoxide to a level of less than about one percent, by volume, or below. An example of a suitable low temperature modifying catalyst are those that are operable at a temperature in a range of between about 150°C and about 300°C. Preferably, the low temperature modifying catalyst includes cupric oxide ( $CuO$ ) and zinc oxide ( $ZnO$ ). Other types of low temperature shift catalysts include copper supported on other transition metal oxides like zirconia, zinc supported on transition metal oxides or refractory supports like silica or alumina, supported platinum, supported rhodium, supported palladium, supported rhodium and supported gold. Low temperature shift zone catalyst 84 is held in place by lower perforated plate 86 and upper perforated plate 88. Gaseous products from cooling zone 72 can pass through perforated plate 86 through low temperature shift zone 76 through upper perforated plate 88. Exit zone 90 is above low temperature shift zone 76 and has reformer exit 92.

[0021] In the method for converting hydrocarbon fuel into hydrogen gas, an oxygen-containing gas, such as air, is directed from oxygen source 42 through conduit 35 to inlet housing 33 to oxygen gas zone 40 into first end inlet 36 of helical tube 32. Reformer 10 can operate at a pressure in the range of between about 0 and 500

psig. The oxygen-containing gas, such as air, is preheated to a temperature of about 450°C. In a preferred embodiment, air has a velocity of greater than about 40 meters per second.

- 5 [0022] A suitable hydrocarbon or alcohol vapor is directed from fuel source 48 through fuel tube 50 to fuel inlet 46. Examples of suitable hydrocarbon fuels include gasoline, JP-8, methanol, ethanol, kerosene and other suitable hydrocarbons typically used in reformers. Gaseous hydrocarbons, such as methane or propane, can also be used. Steam is directed from steam source 54 through steam tube 56 to first steam inlet 52. Steam has a temperature in the range between about 100 and about 150°C. The air, steam and hydrocarbon fuel are fed at rates sufficient to mix within helical tube 32 and spontaneously partially oxidize as the mixture enters partial oxidation zone 24 through first vessel inlet 20 to form a heated reformate stream that includes carbon monoxide and hydrogen gas. In a preferred embodiment, oxygen-containing gas is tangentially directed around the interior of partial oxidation zone 24, which is an empty chamber. In partial oxidation zone 24, the reformate products can include methane, hydrogen gas, water and carbon monoxide. Partial oxidation zone 24 has a preferred temperature in the range of between about 950°C and about 1150°C. A heavier fuel is preferentially run at the higher end of the temperature range while a lighter fuel is run at a lower end of the temperature range.
- 10 [0023] From partial oxidation zone 24, reformate products are directed through perforated plate 30 to steam reforming zone 26. In steam reforming zone 26, the remaining hydrocarbon vapor in the heated reformate stream from partial oxidation zone 24 is steam reformed in the presence of steam reforming catalyst 28 into hydrogen gas and carbon monoxide. Steam reforming zone 26 typically has a temperature in the range of between about 700 and 900°C. The partial oxidation reaction provides sufficient heat to provide heat to helical tube 32 to preheat the air and other contents of helical tube 32 and also provide heat to the steam reforming step. The hydrocarbon fuel is burned partly in partial oxidation zone 24 and the remainder of the fuel with the steam is mixed with the partial oxidation zone combustion products for steam reforming and hydrocarbon shifting to carbon monoxide and hydrogen gas in the presence of steam reforming catalyst 28. The heated reformate stream exiting from steam reforming zone 26 has a temperature of between about 700°C and about 900°C. The heated reformate stream is directed between first vessel 18 and second vessel 58 and around the exterior of helical tube 32, whereby the heated reformate stream is cooled by heating the contents of helical tube 32 and also the first vessel 18 and second vessel 56.
- 15 [0024] Heated reformate stream exits second vessel outlet 62 to flow distribution zone 63, where it has been cooled to a temperature of between about 300°C and
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about 600°C and is directed through perforated plate 68 to high temperature shift zone 64 where essentially all of the carbon monoxide is removed or reduced by contacting the heated reformate stream with high temperature shift catalyst 66 at a temperature in the range of between about 300°C and 600°C. High-temperature shift zone 64 operates adiabatically to reduce the carbon monoxide levels with modest temperature rise. In one embodiment, heated reformate stream entering high temperature shift zone 64 has about fourteen to seventeen percent carbon monoxide, by volume, and exits high temperature shift zone 64 with about two to four percent carbon monoxide, by volume.

[0025] The high temperature shift zone treated reformate stream is directed through sulfur removal zone 71 where the hydrogen sulfide content of the stream is reduced to a concentration of less than about one part per million. From sulfur removal zone 71, the reformate is directed to cooling zone 72 where the stream contacts the vertical fins 74 and cooling tubes 78 to lower the temperature of the stream to between about 150°C and about 300°C because low temperature shift catalyst 84 is temperature sensitive and could possibly sinter at a temperature of above about 300°C. Cooling zone 72 cools high temperature reformate gas for low temperature shift zone 76. Cooling zone tubes 78 operate continuously flooded to allow accurate and maximum steam side heat transfer, to reduce fouling and corrosion to allow use of contaminated water, and to achieve a constant wall minimum temperature.

[0026] Reformate stream is directed through perforated plate 86 to low temperature shift reaction zone 76 where the reformate stream contacts low temperature shift catalyst 84 converting at least a portion of the remaining carbon monoxide gas of the reformate stream to carbon dioxide by low temperature shift reaction to form product stream. Low temperature shift reaction zone 76 operates adiabatically to reduce the remainder of the carbon monoxide to trace levels with modest catalyst temperature rise. The resulting gas product stream exits low temperature shift reaction zone 76 through perforated plate 88 to exit gas zone 90 to reformer exit 92. The exiting product stream can have a composition of about 40% hydrogen gas and less than one percent carbon monoxide on a wet volume basis.

#### Equivalents

[0027] Those skilled in the art will recognize or be able to ascertain using no more than routine experimentation, many equivalents to the specific embodiments of the invention described specifically herein. Such equivalents are intended to be encompassed in the scope of the claims.

#### Claims

1. A reformer for converting a hydrocarbon fuel into hydrogen gas and carbon dioxide comprising:
  - 5 a first vessel (18) having a first portion defining a partial oxidation reaction zone (24) and a first vessel inlet (20) adapted to direct reactants including oxygen-containing gas and unburned fuel tangentially into the first portion of the first vessel (18); and,
  - 10 a first vessel outlet (22) disposed downstream of the partial oxidation reaction zone (24).
- 15 2. The reformer of claim 1 wherein the partial oxidation reaction zone (24) is adapted to permit reactants to flow therethrough in a substantially helical manner in response to tangentially delivered unburned fuel.
- 20 3. The reformer of claim 1 wherein the first vessel (18) includes means to ignite the reactants.
4. The reformer of claim 1 wherein the partial oxidation reaction zone (24) contains an open interior space without any catalyst.
- 25 5. The reformer of claim 1 further comprising:
  - 30 a helical tube (32) disposed about the first vessel (18) and having a first end (34) for connection to an oxygen source (42) and a second end (44) for connection to the first vessel inlet (20). the helical tube (32) adapted to conduct the oxygen-containing gas from the oxygen source (42) to the first vessel (18); and,
  - 35 a second vessel (58) having a second vessel inlet (60) and a second vessel outlet (62), the second vessel (58) annularly disposed about the first vessel (18) and adapted to direct product gases from the first vessel (18) through the second vessel (58), wherein the helical tube (32) is interposed between the first vessel (18) and the second vessel (58).
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  - 45 6. The reformer of claim 1 wherein the first vessel (18) has a second portion disposed downstream of the first portion and defining a steam reforming reaction zone (26), and the first vessel outlet (22) communicates with the steam reforming reaction zone (26).
  - 50 7. The reformer of claim 6 wherein the first vessel (18) includes a perforated barrier (30) interposed between the first and second portions.
  - 55 8. The reformer of claim 6 further comprising:
    - 8 a helical tube (32) disposed about the first vessel (18) and having a first end (34) for connec-

tion to an oxygen source (42) and a second end (44) for connection to the first vessel inlet (20), the helical tube (32) adapted to conduct the oxygen-containing gas from the oxygen source (42) to the first vessel (18); and. a second vessel (58) having a second vessel inlet (60) and a second vessel outlet (62), the second vessel (58) annularly disposed about the first vessel (18) and adapted to direct product gases from the first vessel (18) through the second vessel (58), wherein the helical tube (32) is interposed between the first vessel (18) and the second vessel (58).

9. The reformer of claim 8 wherein the oxygen source (42) is adapted to supply air. 15

10. The reformer of claim 8 further comprising a conduit (53) communicating with a water source and the second vessel (58) and adapted to direct a supply of water to the second vessel (58). 20

11. The reformer of claim 8 wherein the helical tube (32) is connected to a fuel source (48) and is adapted to permit fuel from the fuel source (48) to mix with the oxygen-containing gas within the helical tube (32). 25

12. The reformer of claim 11 wherein the helical tube (32) is connected to a steam source (54) and is adapted to permit steam from the steam source (54) to mix with the fuel and the oxygen-containing gas within the helical tube (32). 30

13. The reformer of claim 6 wherein the steam reforming reaction zone (26) contains a first catalyst (28). 35

14. The reformer of claim 13 wherein the first catalyst (28) includes nickel. 40

15. The reformer of claim 6 further comprising a third vessel annularly disposed about the second vessel (58), the third vessel having a third vessel inlet (63) and a third vessel outlet (90) for directing product gases from the second vessel (58) through the third vessel. 45

16. The reformer of claim 15 wherein the third vessel has a high-temperature shift reaction zone (64) containing a second catalyst (66). 50

17. The reformer of claim 16 wherein the second catalyst (66) includes iron oxide. 55

18. The reformer of claim 15 wherein the third vessel includes a cooling zone (72). 55

19. The reformer of claim 18 wherein the cooling zone (72) contains a cooling tube (78) having a cooling tube inlet (80) adapted for communication with a cooling medium source and a cooling tube outlet (82).

5 20. The reformer of claim 19 wherein the cooling medium source is adapted to direct a liquid-phase cooling medium into the cooling tube (78) and maintain a two-phase mixture throughout substantially an entire length of the cooling tube (78) within the cooling zone (72).

10 21. The reformer of claim 19 wherein the cooling tube (78) is helically disposed within the cooling zone (72).

22. The reformer of claim 18 wherein the third vessel has a low-temperature shift reaction zone (76) including a third catalyst (84) and is disposed downstream of the cooling zone (72).

23. The reformer of claim 22 wherein the third catalyst (84) includes copper.

24. The reformer of claim 22 wherein the third vessel has a high-temperature shift reaction zone (64) disposed upstream of the low-temperature shift reaction zone (76) and the cooling zone (72) is interposed between the high-temperature and low-temperature shift reaction zones (64, 76).

25 25. A method for converting hydrocarbon fuel into hydrogen gas and carbon dioxide within a reformer comprising the steps of:

- a. directing a stream including an oxygen-containing gas adjacent to a first vessel (18) to preheat the oxygen-containing gas by heat transfer from the first vessel (18);
- b. introducing a stream including unburned fuel into the oxygen-containing gas stream to form a mixture including oxygen-containing gas and fuel;
- c. directing the mixture of oxygen-containing gas and fuel tangentially into a partial oxidation reaction zone (24) within the first vessel (18); and,
- d. directing the mixture of oxygen-containing gas and fuel through the partial oxidation reaction zone (24) to produce a heated reformat stream including hydrogen gas and carbon monoxide.

26. The method of claim 25 further comprising the step of directing the mixture of oxygen-containing gas and fuel through the partial oxidation reaction zone (24) in a substantially helical manner.

27. The method of claim 25 further comprising the steps

of introducing steam into the first vessel (18) and directing the reformatate stream from the partial oxidation reaction zone (24) to a steam reforming reaction zone (26) including a first catalyst to produce additional hydrogen gas in the reformatate stream.

28. The method of claim 27 further including the steps of:

directing the stream including the oxygen-containing gas from an oxygen source (42) through a helical tube (32) interposed between the first vessel (18) and a second vessel (58), and into the first vessel (18);

directing the reformatate stream from the steam reforming reaction zone (26) through the second vessel (58), wherein heat energy is transferred from the reformatate stream in the second vessel (58) to the stream including oxygen-containing gas in the helical tube (32).

29. The method of claim 28 further including the step of introducing water into the second vessel (58) from a water source external to the reformer.

30. The method of claim 28 further including the step of introducing the fuel stream and steam into the helical tube (32).

31. The method of claim 27 further including the step of providing a first catalyst (28) including nickel in the steam reforming reaction zone (26).

32. The method of claim 28 further comprising the step of directing the reformatate stream from the second vessel (58) to a third vessel.

33. The method of claim 32 further comprising the step of directing the reformatate stream through a high-temperature shift reaction zone (64) to convert a portion of the carbon monoxide of the reformatate stream into carbon dioxide, wherein the high-temperature shift reaction zone (64) includes a second catalyst (66) and is disposed within the third vessel.

34. The method of claim 33 further providing the step of providing a second catalyst (66) including iron oxide in the high-temperature shift reaction zone (64).

35. The method of claim 32 further comprising the step of directing the reformatate stream through a cooling zone (72) disposed in the third vessel.

36. The method of claim 35 further comprising the steps of radially extending a plurality of fins (74) between an inner wall and outer wall of the third vessel, and directing a cooling medium through a cooling tube (78) disposed within the third vessel to transfer heat from the reformatate stream to the fins (74) and to the cooling medium within the cooling tube (78).

37. The method of claim 36 further comprising the step of maintaining a two-phase mixture of the cooling medium throughout substantially an entire length of the cooling tube (78) within the cooling zone (72).

38. The method of claim 36 wherein the cooling tube (78) is disposed in a helical manner within the cooling zone (72).

39. The method of claim 35 further comprising the step of directing the reformatate stream from the cooling zone (72) to a low-temperature shift reaction zone (76) including a third catalyst (84) and disposed within the third vessel.

40. The method of claim 39 further comprising the step of providing a third catalyst (84) including copper within the low-temperature shift reaction zone (76).

41. The method of claim 35 further comprising the step of directing the reformatate stream through a high-temperature shift reaction zone (64) disposed upstream of the cooling zone (72).

#### Patentansprüche

1. Reformer zum Umwandeln eines Kohlenwasserstoff-Brennstoffs in Wasserstoff und Kohlendioxid, welcher aufweist:

ein erstes Gefäß (18) mit einem ersten Bereich, der eine Teiloxydations-Reaktionszone (24) bildet, und einem Eingang (20) des ersten Gefäßes, welcher Reaktionsmittel enthaltend ein sauerstoffhaltiges Gas und unverbrannten Brennstoff tangential in den ersten Bereich des ersten Gefäßes (18) richtet; und einen Auslaß (22) des ersten Gefäßes, welcher sich stromabwärts der Teiloxydations-Reaktionszone (24) befindet.

2. Reformer nach Anspruch 1, worin die Teiloxydations-Reaktionszone (24) so ausgebildet ist, daß sie das Strömen der Reaktionsmittel durch sie hindurch in einer im Wesentlichen wendelförmigen Weise in Abhängigkeit von dem tangential zugeführten unverbrannten Brennstoff ermöglicht.

3. Reformer nach Anspruch 1, worin das erste Gefäß (18) Mittel enthält, um die Reaktionsmittel zu entzünden.

4. Reformer nach Anspruch 1, worin die Teiloxydations-Reaktionszone (24) einen offenen Innenraum

ohne einen Katalysator enthält.

5. Reformer nach Anspruch 1, welcher weiterhin aufweist:

ein wendelförmiges Rohr (32), das sich um das erste Gefäß (18) befindet und ein erstes Ende (34) zur Verbindung mit einer Sauerstoffquelle (42) und ein zweites Ende (44) zur Verbindung mit dem Einlaß (20) des ersten Gefäßes hat, wobei das wendelförmige Rohr (32) ausgebildet ist, um das sauerstoffhaltige Gas von der Sauerstoffquelle (42) zu dem ersten Gefäß (18) zu führen; und ein zweites Gefäß (58) mit einem Einlaß (60) des zweiten Gefäßes und einem Auslaß (62) des zweiten Gefäßes, wobei das zweite Gefäß (58) ringförmig um das erste Gefäß (18) angeordnet und so ausgebildet ist, daß es Produktgase von dem ersten Gefäß (18) durch das zweite Gefäß (58) leitet, worin das wendelförmige Rohr (32) zwischen dem ersten Gefäß (18) und dem zweiten Gefäß (58) angeordnet ist.

6. Reformer nach Anspruch 1, worin das erste Gefäß (18) einen zweiten Bereich hat, der sich stromabwärts des ersten Bereichs befindet und eine Dampfnachformierungs-Reaktionszone (26) bildet, und der Auslaß (22) des ersten Gefäßes mit der Dampfnachformierungs-Reaktionszone (26) kommuniziert.

7. Reformer nach Anspruch 6, worin das erste Gefäß (18) eine perforierte Barriere (30) enthält, die sich zwischen dem ersten und zweiten Bereich befindet.

8. Reformer nach Anspruch 6, welcher weiterhin aufweist:

ein wendelförmiges Rohr (32), das um das erste Gefäß (18) angeordnet ist und ein erstes Ende (34) zur Verbindung mit einer Sauerstoffquelle (42) und ein zweites Ende (44) zur Verbindung mit dem Einlaß (20) des ersten Gefäßes hat, wobei das wendelförmige Rohr (32) ausgebildet ist, um das sauerstoffhaltige Gas von der Sauerstoffquelle (42) zu dem ersten Gefäß (18) zu leiten; und ein zweites Gefäß (58) mit einem Einlaß (60) des zweiten Gefäßes und einen Auslaß (62) des zweiten Gefäßes, wobei das zweite Gefäß (58) ringförmig um das erste Gefäß (18) angeordnet und so ausgebildet ist, daß es Produktgase von dem ersten Gefäß (18) durch das zweite Gefäß (58) leitet, worin das wendelförmige Rohr (32) zwischen dem ersten Gefäß (18) und dem zweiten Gefäß (58) angeordnet ist.

9. Reformer nach Anspruch 8, worin die Sauerstoffquelle (42) so ausgebildet ist, daß sie Luft liefert.

5 10. Reformer nach Anspruch 8, welcher weiterhin eine Leitung (53) aufweist, die mit einer Wasserquelle und dem zweiten Gefäß (58) kommuniziert und so ausgebildet ist, daß sie dem zweiten Gefäß (58) Wasser zuführt.

10 11. Reformer nach Anspruch 8, worin das wendelförmige Rohr (32) mit einer Brennstoffquelle (48) verbunden und so ausgebildet ist, daß es das Mischen von Brennstoff von der Brennstoffquelle (48) mit dem sauerstoffhaltigen Gas innerhalb des wendelförmigen Rohres (32) ermöglicht.

15 12. Reformer nach Anspruch 11, worin das wendelförmige Rohr (32) mit einer Dampfquelle (54) verbunden und so ausgebildet ist, daß es das Mischen von Dampf von der Dampfquelle (54) mit dem Brennstoff und dem sauerstoffhaltigen Gas innerhalb des wendelförmigen Rohres (32) ermöglicht.

20 13. Reformer nach Anspruch 6, worin die Dampfnachformierungs-Reaktionszone (26) einen ersten Katalysator (28) enthält.

25 14. Reformer nach Anspruch 13, worin der erste Katalysator (28) Nickel enthält.

15 15. Reformer nach Anspruch 6, welcher weiterhin ein drittes Gefäß aufweist, das ringförmig um das zweite Gefäß (58) angeordnet ist, wobei das dritte Gefäß einen Einlaß (63) des dritten Gefäßes und einen Auslaß (90) des dritten Gefäßes hat, um Produktgase von dem zweiten Gefäß (58) durch das dritte Gefäß zu leiten.

30 16. Reformer nach Anspruch 15, worin das dritte Gefäß eine Hochtemperaturverschleubungs-Reaktionszone (64) enthaltend einen zweiten Katalysator (66) hat.

35 17. Reformer nach Anspruch 16, worin der zweite Katalysator (66) Eisenoxid enthält.

40 18. Reformer nach Anspruch 15, worin das dritte Gefäß eine Kühlzone (72) enthaltet.

45 19. Reformer nach Anspruch 18, worin die Kühlzone (72) ein Kühlrohr (78) mit einem Einlaß (80) für das Kühlrohr, das zur Kommunikation mit einer Kühlmediumquelle ausgebildet ist, und einem Auslaß (82) für das Kühlrohr enthält.

50 20. Reformer nach Anspruch 19, worin die Kühlmediumquelle ausgebildet ist.

umquelle so ausgebildet ist, daß sie ein Flüssigphasen-Kühlmedium in das Kühlrohr (78) leitet und eine Zweiphasenmischung über im Wesentlichen die gesamte Länge des Kühlrohres (78) innerhalb der Kühlzone (72) aufrecht erhält.

21. Reformer nach Anspruch 19, worin das Kühlrohr (78) wendelförmig innerhalb der Kühlzone (72) angeordnet ist.

22. Reformer nach Anspruch 18, worin das dritte Gefäß eine Niedrigtemperaturverschiebungs-Reaktionszone (76) enthaltend einen dritten Katalysator (84) hat und sich stromabwärts der Kühlzone (72) befindet.

23. Reformer nach Anspruch 22, worin der dritte Katalysator (84) Kupfer enthält.

24. Reformer nach Anspruch 22, worin das dritte Gefäß eine Hochtemperaturverschiebungs-Reaktionszone (64) hat, die sich stromaufwärts der Niedrigtemperaturverschiebungs-Reaktionszone (76) befindet, und die Kühlzone (72) zwischen der Hochtemperatur- und der Niedrigtemperaturverschiebungs-Reaktionszone (64, 76) angeordnet ist.

25. Verfahren zum Umwandeln von Kohlenwasserstoff-Brennstoff in Wasserstoff und Kohlendioxid innerhalb eines Reformers, welches die Schritte aufweist:

- Leiten eines Stromes enthaltend ein sauerstoffhaltiges Gas benachbart zu einem ersten Gefäß (18), um das sauerstoffhaltige Gas durch Wärmeübertragung von dem ersten Gefäß (18) vorzuwärmen;
- Einführen eines Stroms enthaltend nichtverbrannten Brennstoff in den Strom des sauerstoffhaltigen Gases, um eine Mischung enthaltend sauerstoffhaltiges Gas und Brennstoff zu bilden;
- Leiten der Mischung aus sauerstoffhaltigem Gas und Brennstoff tangential in eine Teiloxydations-Reaktionszone (24) innerhalb des ersten Gefäßes (18); und
- Leiten der Mischung aus sauerstoffhaltigem Gas und Brennstoff durch die Teiloxydations-Reaktionszone (24), um einen erwärmten nachformierten Strom enthaltend Wasserstoff und Kohlenmonoxid zu erzeugen.

26. Verfahren nach Anspruch 25, welches weiterhin den Schritt des Leitens der Mischung aus sauerstoffhaltigem Gas und Brennstoff durch die Teiloxydations-Reaktionszone (24) in einer im Wesentlichen wendelförmigen Weise aufweist.

27. Verfahren nach Anspruch 25, welches weiterhin die Schritte der Einführung von Dampf in das erste Gefäß (18) und des Leitens des nachformierten Stroms von der Teiloxydations-Reaktionszone (24) in eine Dampfnachformierungs-Reaktionszone (26) enthaltend einen ersten Katalysator, um zusätzlichen Wasserstoff in dem nachformierten Strom zu erzeugen, aufweist.

28. Verfahren nach Anspruch 27, welches weiterhin die Schritte aufweist:  
Leiten des Stroms enthaltend das sauerstoffhaltige Gas von einer Sauerstoffquelle (42) durch ein wendelförmiges Rohr (32), das zwischen dem ersten Gefäß (18) und einem zweiten Gefäß (58) angeordnet ist, und in das erste Gefäß (18); Leiten des nachformierten Stroms von der Dampfnachformierungs-Reaktionszone (26) durch das zweite Gefäß (58), worin Wärmeenergie von dem nachformierten Strom in dem zweiten Gefäß (58) auf den Strom enthaltend sauerstoffhaltiges Gas in dem wendelförmigen Rohr (32) übertragen wird.

29. Verfahren nach Anspruch 28, weiterhin enthaltend den Schritt des Einführens von Wasser in das zweite Gefäß (58) von einer außerhalb des Reformers befindlichen Wasserquelle.

30. Verfahren nach Anspruch 28, weiterhin enthaltend den Schritt des Einführens des Brennstoffstroms und Dampfes in das wendelförmige Rohr (32).

31. Verfahren nach Anspruch 27, weiterhin enthaltend den Schritt des Vorsehens eines ersten Katalysators (28) enthaltend Nickel in der Dampfnachformierungs-Reaktionszone (26).

32. Verfahren nach Anspruch 28, welches weiterhin den Schritt des Leitens des nachformierten Stroms aus dem zweiten Gefäß (58) in ein drittes Gefäß aufweist.

33. Verfahren nach Anspruch 32, welches weiterhin den Schritt des Leitens des nachformierten Stroms durch eine Hochtemperaturverschiebungs-Reaktionszone (64) aufweist, um einen Teil des Kohlenmonoxids des nachformierten Stroms in Kohlendioxid umzuwandeln, worin die Hochtemperaturverschiebungs-Reaktionszone (64) einen zweiten Katalysator (66) enthält und sich innerhalb des dritten Gefäßes befindet.

34. Verfahren nach Anspruch 33, welches weiterhin den Schritt des Vorsehens eines zweiten Katalysators (66) enthaltend Eisenoxid in der Hochtemperaturverschiebungs-Reaktionszone (64) aufweist.

turverschiebungs-Reaktionszone (64) aufweist.

35. Verfahren nach Anspruch 32, welches weiterhin den Schritt des Leitens des nachformierten Stroms durch eine in dem dritten Gefäß angeordnete Kühlzone (72) aufweist. 5

36. Verfahren nach Anspruch 35, welches weiterhin die Schritte der radialen Verlängerung mehrerer Rippen (74) zwischen einer Innenwand und einer Außenwand des dritten Gefäßes und des Leitens eines Kühlmediums durch ein Kühlrohr (78), das sich innerhalb des dritten Gefäßes befindet, um Wärme von dem nachformierten Strom zu den Rippen (74) und zu dem Kühlmedium innerhalb des Kühlrohres (78) zu übertragen, aufweist. 10

37. Verfahren nach Anspruch 36, welches weiterhin den Schritt der Aufrechterhaltung einer Zweiphasenmischung des Kühlmediums über im Wesentlichen die gesamte Länge des Kühlrohres (78) innerhalb der Kühlzone (72) aufweist. 15

38. Verfahren nach Anspruch 36, worin das Kühlrohr (78) in einer wendelförmigen Weise innerhalb der Kühlzone (72) angeordnet ist. 20

39. Verfahren nach Anspruch 35, welches weiterhin den Schritt des Leitens des nachformierten Stroms von der Kühlzone (72) zu einer Niedrigtemperaturverschiebungs-Reaktionszone (76), die einen dritten Katalysator (84) enthält und sich innerhalb des dritten Gefäßes befindet, aufweist. 25

40. Verfahren nach Anspruch 39, welches weiterhin den Schritt des Vorsehens eines dritten Katalysators (84) enthaltend Kupfer innerhalb der Niedrigtemperaturverschiebungs-Reaktionszone (76) aufweist. 30

41. Verfahren nach Anspruch 35, welches weiterhin den Schritt des Leitens des nachformierten Stroms durch eine Hochtemperaturverschiebungs-Reaktionszone (64), die sich stromaufwärts der Kühlzone (74) befindet, aufweist. 35

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comportant en outre :

un tube hélicoïdal (32) disposé autour du premier réceptacle (18) et ayant une première extrémité (34) destinée à être raccordée à une source d'oxygène (42) et une seconde extrémité (44) destinée à être raccordée à l'entrée (20) du premier réceptacle, le tube hélicoïdal (32) étant destiné à conduire le gaz contenant l'oxygène de la source d'oxygène (42) au premier réceptacle (18), et un second réceptacle (58) ayant une entrée (60) et une sortie (62) de second réceptacle, le second réceptacle (58) étant disposé annulement autour du premier réceptacle (18) et étant destiné à diriger les gaz produits du premier réceptacle (18) dans le second réceptacle (58), le tube hélicoïdal (32) étant disposé entre le premier réceptacle (18) et le second réceptacle (58).

9. Appareil de reformage selon la revendication 8, dans lequel la source d'oxygène (42) est destinée à transmettre de l'air.
10. Appareil de reformage selon la revendication 8, comportant en outre un conduit (53) qui communique avec une source d'eau et le second réceptacle (58) et qui est destiné à diriger une réserve d'eau vers le second réceptacle (58).
11. Appareil de reformage selon la revendication 8, dans lequel le tube hélicoïdal (32) est raccordé à une source de combustible (48) et est destiné à permettre le mélange du combustible de la source de combustible (48) avec le gaz contenant de l'oxygène à l'intérieur du tube hélicoïdal (32).
12. Appareil de reformage selon la revendication 11, dans lequel le tube hélicoïdal (32) est raccordé à une source de vapeur d'eau (54) et est destiné à permettre à la vapeur d'eau de la source de vapeur d'eau (54) de se mélanger au combustible et au gaz contenant l'oxygène dans le tube hélicoïdal (32).
13. Appareil de reformage selon la revendication 6, dans lequel la zone (26) de réaction par reformage à la vapeur d'eau contient un premier catalyseur (28).
14. Appareil de reformage selon la revendication 13, dans lequel le premier catalyseur (28) contient du nickel.
15. Appareil de reformage selon la revendication 6, comportant en outre un troisième réceptacle disposé annulement autour du second réceptacle (58), le troisième réceptacle ayant une entrée (63),

et une sortie (90) de troisième réceptacle destinée à diriger les produits gazeux du second réceptacle (58) dans le troisième réceptacle.

- 5 16. Appareil de reformage selon la revendication 15, dans lequel le troisième réceptacle a une zone (64) de réaction par transposition à haute température contenant un second catalyseur (66).
- 10 17. Appareil de reformage selon la revendication 16, dans lequel le second catalyseur (66) contient de l'oxyde de fer.
- 15 18. Appareil de reformage selon la revendication 15, dans lequel le troisième réceptacle contient une zone de refroidissement (72).
- 20 19. Appareil de reformage selon la revendication 18, dans lequel la zone de refroidissement (72) contient un tube de refroidissement (78) ayant une entrée (80) de tube de refroidissement destinée à communiquer avec une source d'un fluide de refroidissement et une sortie (82) de tube de refroidissement.
- 25 20. Appareil de reformage selon la revendication 19, dans lequel la source de fluide de refroidissement est destinée à diriger un fluide de refroidissement en phase liquide dans le tube de refroidissement (78) et à maintenir un mélange biphasé pratiquement dans toute la longueur du tube de refroidissement (78) dans la zone de refroidissement (72).
- 30 21. Appareil de reformage selon la revendication 19, dans lequel le tube de refroidissement (78) est disposé en hélice dans la zone de refroidissement (72).
- 35 22. Appareil de reformage selon la revendication 18, dans lequel le troisième réceptacle a une zone (76) de réaction par transposition à basse température contenant un troisième catalyseur (84) et disposé en aval de la zone de refroidissement (72).
- 40 23. Appareil de reformage selon la revendication 22, dans lequel le troisième catalyseur (84) contient du cuivre.
- 45 24. Appareil de reformage selon la revendication 22, dans lequel le troisième réceptacle a une zone (84) de réaction par transposition à haute température disposée en amont de la zone (76) de réaction par transposition à basse température et la zone de refroidissement (72) est disposée entre les zones (64, 76) de réaction par transposition à haute température et à basse température.
- 50 25. Procédé de transformation d'un combustible hydrocarboné en hydrogène gazeux et anhydride carbo-

nique dans un appareil de reformage, comprenant les étapes suivantes :

- a) la direction d'un courant contenant un gaz qui contient de l'oxygène près d'un premier réceptacle (18) afin que le gaz contenant de l'oxygène soit préchauffé par transfert de chaleur depuis le premier réceptacle (18),
- b) l'introduction d'un courant contenant un combustible imbrûlé dans le courant de gaz contenant de l'oxygène pour la formation d'un mélange contenant le gaz contenant l'oxygène et du combustible,
- c) la direction du mélange du gaz contenant de l'oxygène et du combustible tangentiellement dans une zone (24) de réaction par oxydation partielle à l'intérieur du premier réceptacle (18), et
- d) la direction du mélange du gaz contenant de l'oxygène et du combustible dans la zone (24) de réaction par oxydation partielle pour la production d'un courant de reformat chauffé contenant de l'hydrogène gazeux et de l'anhydride carbonique.

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26. Procédé selon la revendication 25, comprenant en outre une étape de direction du mélange du gaz contenant de l'oxygène et du combustible dans la zone (24) de réaction par oxydation partielle pratiquement suivant un trajet hélicoïdal.

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27. Procédé selon la revendication 25, comprenant en outre des étapes d'introduction de vapeur d'eau dans le premier réceptacle (18), et de direction du courant de reformat de la zone (24) de réaction par oxydation partielle dans une zone (26) de réaction par reformage à la vapeur d'eau contenant un premier catalyseur pour la production d'hydrogène gazeux supplémentaire dans le courant de reformat.

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28. Procédé selon la revendication 27, comprenant en outre les étapes suivantes :

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la direction du courant contenant le gaz contenant de l'oxygène provenant d'une source d'oxygène (42) dans un tube hélicoïdal (32), disposé entre le premier réceptacle (18) et un second réceptacle (58), vers le premier réceptacle (18), et  
 la direction du courant de reformat provenant de la zone (26) de réaction par reformage à la vapeur d'eau dans le second réceptacle (58), dans lequel de l'énergie thermique est transférée du courant de reformat présent dans le second réceptacle (58) au courant contenant le gaz contenant l'oxygène dans le tube hélicoïdal (32).

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29. Procédé selon la revendication 28, comprenant en outre une étape d'introduction d'eau dans le second réceptacle (58) depuis une source d'eau extérieure à l'appareil de reformage.

30. Procédé selon la revendication 28, comprenant en outre une étape d'introduction du courant de combustible et de vapeur d'eau dans le tube hélicoïdal (32).

31. Procédé selon la revendication 27, comprenant en outre une étape de disposition d'un premier catalyseur (28) qui contient du nickel dans la zone (26) de réaction par reformage à la vapeur d'eau.

32. Procédé selon la revendication 28, comprenant en outre une étape de direction du courant de reformat du second réceptacle (58) vers un troisième réceptacle.

33. Procédé selon la revendication 32, comprenant en outre une étape de direction du courant de reformat dans une zone (64) de réaction par transposition à haute température afin qu'une partie de l'oxyde de carbone du courant de reformat se transforme en anhydride carbonique, la zone (64) de réaction par transposition à haute température contenant un second catalyseur (66) et étant disposée dans le troisième réceptacle.

34. Procédé selon la revendication 33, comprenant en outre une étape de disposition d'un second catalyseur (66) contenant de l'oxyde de fer dans la zone (64) de réaction par transposition à haute température.

35. Procédé selon la revendication 32, comprenant en outre une étape de direction du courant de reformat dans une zone de refroidissement (72) placée dans le troisième réceptacle.

36. Procédé selon la revendication 35, comprenant en outre des étapes de disposition de plusieurs ailettes (74) entre une paroi interne et une paroi externe du troisième réceptacle, et de direction d'un fluide de refroidissement par l'intermédiaire d'un tube de refroidissement (78) placé dans le troisième réceptacle afin que de la chaleur soit transférée du courant de reformat aux ailettes (74) et au fluide de refroidissement placé dans le tube de refroidissement (78).

37. Procédé selon la revendication 36, comprenant en outre une étape de maintien d'un mélange biphasé du fluide de refroidissement pratiquement sur toute la longueur du tube de refroidissement (78) à l'intérieur de la zone de refroidissement (72).

38. Procédé selon la revendication 36, dans laquelle le tube de refroidissement (78) est disposé en hélice dans la zone de refroidissement (72).

39. Procédé selon la revendication 35, comprenant en outre une étape de direction du courant de reformage de la zone de refroidissement (72) vers une zone (76) de réaction par transposition à basse température qui contient un troisième catalyseur (84) et qui est disposée dans le troisième réceptacle. 5

40. Procédé selon la revendication 39, comprenant en outre une étape de disposition d'un troisième catalyseur (84) contenant du cuivre (76) de réaction par transposition à basse température. 10

41. Procédé selon la revendication 35, comprenant en outre une étape de direction du courant de reformage dans une zone (64) de réaction par transposition à haute température placée en amont de la zone de refroidissement (72). 15

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